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(21) International Application Number: PCT/US88/04588 (22) International Filing Date: 21 December 1988 (21.12.88) (31) Priority Application Number: 138,722 (32) Priority Date: 23 December 1987 (23.12.87) (33) Priority Country: US (71) Applicant: AVERY INTERNATIONAL CORPORATION [US/US]; 150 North Orange Grove Boulevard, Pasadena, CA 91103 (US). (72) Inventors: MALLYA, Prakash ; 926 Crestview Drive, Pasadena, CA 91107 (US). SMITH, Colin ; 818 Green Street, Glendale, CA 91205 (US). PLAMTHOTTAM, Sebastian, S. ; 280 E. Del Mar #303, Pasadena, CA 91101 (US).		(74) Agent: GRINNELL, John, P.; Christie, Parker & Hale, P.O. Box 7068, Pasadena, CA 91109-7068 (US). (81) Designated States: AT (European patent), AU, BE (European patent), CH (European patent), DE (European patent), FR (European patent), GB (European patent), IT (European patent), JP, KR, LU (European patent), NL (European patent), SE (European patent). Published <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
(54) Title: HIGH PERFORMANCE PRESSURE-SENSITIVE ADHESIVE POLYMERS		
(57) Abstract Pressure-sensitive copolymers based on acrylic monomers are provided with high adhesive performance characteristics by the inclusion of a synergistic amount of an N-vinyl lactam monomer and a glycidyl monomer with the bulk of the monomers being an alkyl acrylate and/or methacrylate esters.		

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HIGH PERFORMANCE PRESSURE-SENSITIVE
ADHESIVE POLYMERS

BACKGROUND OF THE INVENTION

15 The present invention is directed to pressure-sensitive adhesive compositions that exhibit high adhesion to high energy surfaces such as aluminum and possess high tack and outstanding holding power at elevated temperatures. Two key monomers are used in combination in the pressure-sensitive adhesives of this invention to impart unique properties to the resulting polymers. The monomers are at least one glycidyl monomer in combination with at least one N-vinyl lactam.

The use of glycidyl monomers in pressure-sensitive adhesive has been disclosed in the art.

25 U.S. Patent 3,284,423 discloses creep-resistant pressure-sensitive adhesive compositions comprising 35-75% by weight alkyl acrylate esters containing 6-15 carbon atoms, 10-60% lower alkyl acrylate, 0.1-10% by weight of an ethylenically unsaturated carboxylic acid and 0.1-10% by weight glycidyl ester.

30 U.S. Patent 3,893,982 discloses an interpolymer comprising 0.1-15% parts of an ethylenically unsaturated carboxylic acid, 0.1-2% parts of a glycidyl monomer, 35-84.9% parts of an alkyl acrylate or methacrylate and optionally a monomer selected from the group consisting

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1 of alpha-olefins containing 2-10 carbon atoms, vinyl
esters of alkanolic acids containing 3-10 carbon atoms,
ethyl and methyl esters of acrylic and methacrylic
acids, acrylonitrile, methacrylonitrile, styrene and
5 vinyl chloride where the polymer has a weight average
molecular weight in the range of 10,000 - 500,000 and
between 0.01 and 1 parts by weight per 100 parts of the
copolymer of 1,3-bis(dimethylamino)-2-hydroxypropane to
cause the cure of the epoxy group.

10 The art has also taught the use of N-vinyl lactams
in polymers.

U.S. Patent 3,728,148 discloses a pressure-
sensitive adhesive for electrical insulating
applications comprising of a copolymer of 65-90% by
15 weight of an alkyl acrylate ester, 10-30 by weight of a
N-vinyl lactam and 0-20% by weight of a modifying
monomer which is copolymerizable with the above. Acidic
monomers and amides are excluded as they are claimed to
cause undesirable corrosion.

20 U.S. Patent 4,181,752 discloses an interpolymer
containing 87% by weight isooctyl acrylate, 8% by weight
vinyl pyrrolidone, 3% by weight acrylic acid and 2% by
weight acrylamide.

25 U.S. Patent 4,364,972 discloses a pressure-
sensitive adhesive tape made by copolymerizing an alkyl
acrylate ester with 15 to 50 parts by weight of vinyl
pyrrolidone and having a K-value greater than 100 and
when crosslinked has a gel-swell in ethyl acetate in
excess of 600%. Advantages claimed are good adhesion to
30 automotive paints, rubber and plastic foam layers.

U.S. Patent 4,310,509 discloses a 90/10 2-ethyl
hexyl acrylate/vinyl pyrrolidone copolymer for making a
pressure sensitive adherent for complexing with iodine
for anti-microbial activity.

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1 European Patent Application 130080 discloses an
emulsion polymerized pressure-sensitive adhesive
comprising 2-20% by weight N-vinyl lactam and an alkyl
acrylate ester. The claimed use is good adhesion to
5 skin under hot and humid conditions.

U.S. Patent 4,370,380 is directed to a blend of two
polymers. One is a copolymer of 88-99% by weight of an
alkyl acrylate ester with 1-12% by weight of a
carboxylic acid with a glass transition temperature (Tg)
10 of less than 0°C. The second polymer is either a homo
or a copolymer of N-vinyl lactam with a Tg of 20-150°C.
The blend ratio is 70-99% by weight of the first polymer
with 1-30% by weight of the second polymer. The
resultant pressure-sensitive adhesive is disclosed to
15 have moisture permeability.

U.S. Patent 4,150,197 discloses a water vapor
permeable pressure-sensitive adhesive comprising a
copolymer of 79-89% of butyl acrylate, 10-20% by weight
of N-vinyl lactam and 1-5% by weight of an acidic
20 comonomer.

None of the patents or applications discloses a
copolymer containing both a glycidyl monomer and N-
vinyl lactam monomer.

25 SUMMARY OF THE INVENTION

It has now been found that, as part of an acrylic
and/or methacrylic ester based polymer system, a
glycidyl monomer and a N-vinyl lactam monomer
synergistically act to provide unusually high adhesion
30 to high energy surfaces such as aluminum and stainless
steel. The pressure-sensitive adhesives of the
invention are formed of copolymers containing
essentially no cross-linking when polymerized and which
contain on a copolymerized basis from about 0.1 to about
35 2% by weight of glycidyl monomer, about 1% to about 20%

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1 by weight, preferably from about 1 to about 10% by
weight of an N-vinyl lactam monomer, from 0 to about 15%
by weight of an ethylenically unsaturated carboxylic
acid, from about 55 to about 85% by weight an alkyl
5 acrylate or methacrylate ester containing from 4 to
about 12 carbon atoms in the alkyl group, from 0 to 35%
by weight of an alkyl acrylate or methacrylate ester
containing less than 4 carbon atoms in the alkyl group,
and optionally, one or more other monomers employed to
10 tailor polymer properties, such as glass transition
temperature, to end use applications.

Such monomers include polystyryl ethyl
methacrylate, acetoacetoxy ethyl methacrylate, styrene,
alpha olefins, and vinyl esters of alkanolic acids
15 containing greater than 3 carbon atoms and mixtures
thereof. Modifying monomer content can range from 0 to
about 35% by weight of the total monomers.

The ratio of monomers is selected to provide a
copolymer with glass transition temperature of less than
20 about -15°C and a weight average molecular weight of at
least about 200,000, preferably from about 200,000 to
about 500,000 as determined by size exclusion
chromatography using polystyrene for calibration.
Polymers of the instant invention may be synthesized by
25 solution, emulsion and bulk polymerization. It is
presently preferred that they be formed by solution
polymerization. Polymers are cross-linked to the
desired extent, prior to use, using heat, ionic
additives, actinic or electron beam radiation and the
30 like.

The novel polymers on cross-linking exhibit
excellent adhesion to high energy surfaces such as
aluminum as evidenced by increased peel adhesion with
dwell and superior rivet performance as evidenced by
35 reduced tenting as described herein.

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1 THE DRAWINGS:

 FIG. 1 graphically compares 180° peel adhesion on
alodine aluminum panels of two adhesives, one which
contains both a glycidyl monomer and a lactam monomer to
5 one containing only a glycidyl monomer.

 FIG. 2 compares 180° peel on stainless steel as a
function of dwell at two temperature conditions, room
temperature and 120°C for compositions of the invention
to controls.

10 The room temperature dwell was for 20 minutes and
120°C dwell was for 30 minutes. After dwelling for 30
minutes at 120°C, the adhesive on the substrate was
allowed to equilibrate to room temperature and 180° peel
determined.

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1 DETAILED DESCRIPTION

Pressure-sensitive adhesive polymers of the instant invention are prepared by copolymerizing a mixture of monomers comprising from about 55 to about 85% by weight of an alkyl acrylate and/or methacrylate ester containing 4 to about 12 carbon atoms in the alkyl group; from about 0.01 to about 2% by weight of a glycidyl monomer; from about 1 to about 20% by weight, preferably from 1 to about 10% by weight of an N-vinyl lactam monomer; from 0 to 15% by weight, preferably from about 5 to about 13% by weight of an unsaturated carboxylic acid; from 0 to about 35% by weight of an alkyl acrylate and/or methacrylate ester containing less than 4 carbon atoms in the alkyl group and optionally from about 0 to 33% by weight of one or more other comonomers to provide a balance of desirable polymer properties such as glass transition temperature. The precise ratio of the monomers is selected to give a polymer whose glass transition temperature is lower than about -15°C. The polymers of the instant invention have a weight average molecular weight of at least about 200,000, preferably from about 200,000 to about 500,000 as determined by size exclusion chromatography using polystyrene as the calibrator.

25 The alkyl acrylate and methacrylate esters containing 4 to about 12 carbon atoms in the alkyl group useful in forming the polymers of the instant invention include without limitation 2-ethyl hexyl acrylate, isooctyl acrylate, butyl acrylate, sec-butyl acrylate, methyl butyl acrylate, 4-methyl-2-pentyl acrylate, isodecyl methacrylate and the like and mixtures thereof. Isooctyl acrylate and 2-ethyl hexyl acrylate are presently preferred.

35 The glycidyl monomers are glycidyl acrylate, glycidyl methacrylate, allyl glycidyl ether and mixtures

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1 thereof. The presently preferred glycidyl monomer is glycidyl methacrylate.

 The N-vinyl lactams monomers which may be used include N-vinyl pyrrolidone, N-vinyl caprolactam, 1-
5 vinyl-2-piperidone, 1-vinyl-5-methyl-2-pyrrolidone, and the like, N-vinyl pyrrolidone is presently preferred.

 Ethylenically unsaturated carboxylic acids include acrylic acid, methacrylic acid, fumaric acid, and the like.

10 Alkyl acrylate and methacrylate esters containing less than 4 carbon atoms in the alkyl group include methyl acrylate, ethyl acrylate, methyl methacrylate and the like. Methyl acrylate is presently preferred.

 Other monomers which can be included are polystyryl
15 ethyl methacrylate, acetoacetoxy ethyl methacrylate, alpha olefins such as ethylene and propylene and vinyl esters of alkanolic acids containing more than three carbon atoms as well as mixtures thereof. Such monomer concentrations are in the range from 0 to about 35
20 percent by weight of the total monomers.

 The copolymers may be synthesized using solution emulsion, and batch polymerization techniques. It is presently preferred to prepare the copolymers in solution using a mixture of solvents. The present
25 preferred solution polymerization involves the use of blends of ethyl acetate and hexane or ethyl acetate and acetone. The ratio of solvents are adjusted to provide a reflux temperature of from about 68°C to about 78°C. Solids content during polymerization may typically range
30 from about 40% to about 60% in order to achieve the desired weight average molecular weight, and yet achieve viscosities that are manageable in the reactor. Reaction occurs in the presence of free-radical initiators, preferably of the azo type, for example,
35 2,2'-azobis (isobutyronitrile). The polymers formed are

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1 solvent soluble polymers with essentially no cross-
linking. To this end, the glycidyl monomer is
preferably limited to 2% by weight of the total monomers
to avoid the possibility of cross-linking, by opening of
5 the oxirane group, during polymerization or during
aging. Polymers can, as desired, be post-polymerization
cross-linked using heat, actinic or electron beam
radiation and the like.

The unique characteristics of the cross-linked
10 pressure-sensitive adhesive copolymers of the instant
invention is a dramatic adhesion to high energy
surfaces, such to aluminum and stainless steel, as seen
by increased peel adhesion values with dwell times and
superior rivet performance, as reflected by reduced
15 tenting. Superior performance requires the presence of
both glycidyl and lactam monomers. In the absence of
one, poor adhesion to aluminum has been observed, as
manifested by poor tenting performance (see Table I).
These results are unexpected as a primary purpose in
20 incorporating the N-vinyl lactam is to increase adhesion
to vinyl films and painted surfaces. The purpose of the
glycidyl group is normally to introduce a latent
functional group which could undergo cross-linking at
elevated temperatures under use conditions. This was
25 realized by the dramatic improvement in elevated
temperature performance even as high as 200°C and high
levels of shear adhesion failure temperature (SAFT) of
polymers containing glycidyl methacrylate as established
by Table II. Improved peel adhesion was totally
30 unexpected. While not bound by theory, it is presently
believed that chemical reaction occurs between the epoxy
groups and functional groups such as hydroxyl groups on
the substrate or a complexation reaction occurs between
the epoxy groups and the nitrogen of the lactam, with

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1 ionic groups such as Al+++ on the substrate leading to increased bond strength.

5 An application of the adhesives of the instant invention is the marking of truck panels and the like with an adhesive coated vinyl film. The film is applied over a panel which may be a painted or unpainted aluminum panel fixed with aluminum rivets. The ability of the adhesive to conform to the contour of the rivet and not lift appreciably after application is highly desirable. A tendency to lift away is known as "tenting" and the greater the separation from the rivet the more unsatisfactory is the adhesive.

10 The SAFT test is a test where the adhesive is applied to 0.5" x 1" overlap on stainless steel to which a 4.5 lb. roll force applied. After dwell of 24 hours, this is placed in an oven and a kilogram load is applied under shear conditions and temperature raised from 40°C to 200°C at the rate of 1°C per minute. The failure temperature is recorded as the shear adhesion failure temperature. This is a measure of the cohesive strength of the adhesive or the ability of the adhesive to maintain a bond at elevated temperatures.

20 While not limiting, the following illustrate the invention.

25

Example 1

30 A monomer mixture was made up by mixing 423 g. of 2-ethyl hexyl acrylate, 145 g of methyl acrylate, 3.15 g. of glycidyl methacrylate, 12.6 g of N-vinyl pyrrolidone and 44.1 g of acrylic acid. 157 g. of this mixture was introduced to a 2 liter reactor equipped with a pitched turbine agitator, a reflux condensor and a thermistor. Also added were 73.5 g. of ethyl acetate and 78.76 g of hexane. The contents of the reactor were

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1 heated to reflux and 0.238 g of Vazo 64, manufactured
and sold by duPont in 5.0 g of ethyl acetate was added.
Vigorous reflux started in a short time and the contents
of the reactor were held for 23 minutes. At this time,
5 the remaining monomers were mixed with 537.2 g. of ethyl
acetate, 75.2 g. of hexane and 0.707 g. of Vazo 64 and
added as a single feed mixture over 3.5 hrs. All
through the feed, temperature was maintained to keep
reactor contents under reflux. One hour after end of
10 feed, 0.17 g. Vazo 64 was added in 5 g. ethyl acetate
and held for an additional hour. The percentage of
solids content at the end of reaction was 46.4% and the
viscosity was 23 Pa.s using #4 @ 12 on a Brookfield
viscometer.

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Example 2

A monomer mixture was made up by mixing 453.6 g. of
Isooctyl acrylate, 100.8 g of methyl acrylate, 6.3 g. of
20 glycidyl methacrylate, 25.2 g of N-vinyl caprolactam,
44.1 g of acrylic acid and 0.945 g of Vazo 64. 157.5 g
of this mixture was introduced to the reactor with 78.76
g of hexane, 78.76 g of ethyl acetate and heated to
reflux. Once vigorous reflux initiated, the contents
25 were held for about 12 minutes and the remaining
monomers added along with 537.24 g of ethyl acetate and
75.24 g of hexane as a single feed over 3 hours. Two
hours after the end of feed, the contents were cooled.

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Example 3 - Tenting Test

There was formed as control 1 a polymer containing
65.7% by weight 2-ethyl hexyl acrylate, 27.3% by weight
methyl acrylate and 7% by weight acrylic acid. As
35 Control 2, there was formed a polymer containing 67% by

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1 weight 2-ethyl hexyl acrylate, 24% by weight methyl
acrylate, 7% acrylic acid, and 2% by weight N-vinyl
pyrrolidone. As Control 3, there was formed a polymer
containing 65.7% by weight 2-ethyl hexyl acrylate, 27.2%
5 by weight methyl acrylate, 7% by weight acrylic acid and
0.1% by weight glycidyl methacrylate. As Control 4,
there was used a polymer containing 65% by weight 2-
ethyl hexyl acrylate, 27% by weight methyl acrylate, 7%
by weight acrylic acid and 1% by weight glycidyl
10 methacrylate. These were compared for tenting in a
rivet test to the polymers of Examples 1 and 2. As a
cross-linker in each instance, there was added 0.2 parts
by weight of the polymer of aluminum acetyl acetate.
For the rivet tenting test adhesive was transfer coated
15 from a release liner to a cast vinyl facestock, at a
coat weight of 30 g/m². The results are given in Table
1, wherein the lower the value reported, the less
tenting, i.e., lifting away from the rivet, occurred.

Table 2 compares the polymer of Example 1 electron-
20 beam (EB) cured at a dosage of 30 kiloGray (kGy) and the
polymer of Example 2 also EB cured at a dosage of 30
kGy. Examples 1 and 2 exhibited the best combinations of
shear and tack.

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TABLE 1

	<u>Control/ Example*</u>	<u>Tenting in Rivet Test, mm</u>
5	Control 1	1.42
	Control 2	1.32
	Control 3	1.37
	Control 4	1.25
10	Example 1	0.58
	Example 2	1.1

* All the polymers were cross-linked with 0.2 parts per hundred parts of the polymer of Aluminum acetyl acetate.

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TABLE 2

	<u>Example 1</u>	<u>Example 2</u>
	<u>EB, 30 kGy</u>	<u>EB, 30 kGy</u>
	<u>180° Peel, NM</u>	
20	Stainless Steel, 20' Dwell	572
	Hard PVC	692
		780
	<u>Static Shear, min.</u>	
	20°C/1kg	10000+
25	150°C/1kg	9577
	200°C/1kg	5200+
		3300+
	<u>Loop Tack, N/M</u>	
		600
30	<u>SAFT, °C</u>	270
		200°+
	RT Shear, 0.5 x 0.5 inch overlap, Al Facestock	
	SAFT and ET Shear, 0.5 x 1.0 inch overlap, Al Facestock	
	For SAFT, 24 hr. dwell, rate of heating 1°C/minute	

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Control 5 to 8 and Examples 4 and 5

5 There was compared to the product of Example 2 a
copolymer containing 67 parts by weight isooctyl
acrylate, 25 parts by weight methyl acrylate, 1 part by
weight glycidyl methacrylate, and 7 parts by weight
acrylic acid (Control 5). Control 5 and Example 2
10 copolymers were coated to a level of 50 g/m² onto mylar
and cured at an electron beam level of 30 kGy. A
comparison of 180° peel on alodine aluminum substrate in
Newtons/meter (N/M) as a function of dwell time is shown
in Figure 1 and establishes that the combination of
glycidyl methacrylate and vinyl caprolactam produce
15 unusually high 180° peels, especially after extended
dwell times.

Figure 2 compares 180° peel on stainless steel
substrate as a function of dwell at room temperature for
20 minutes and dwell at 120°C for 30 minutes followed by
20 peel testing at room temperature. All polymers employed
contained 2-ethyl hexyl acrylate, methyl acrylate and
acrylic acid. Control 6 contained no glycidyl
methacrylate or N-vinyl lactam. Control 7 contained 0.1
part glycidyl methacrylate but no N-vinyl lactam.
25 Control 8 contained 1 part by weight glycidyl
methacrylate but no N-vinyl lactam. Example 4 contained
0.1 part by weight glycidyl methacrylate and 2 parts by
weight N-vinyl pyrrolidone. Example 5 contained 0.1
part by weight glycidyl methacrylate and 4 parts by
30 weight N-vinyl caprolactam per hundred parts total
monomer. In each instance, the polymers were EB cured
at a dosage of 30 kGy. The glycidyl methacrylate in
combination and N-vinyl lactam gives better performance
in terms of adhesion to high energy surfaces than the
35 individual constituents of the combination.

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1 WHAT IS CLAIMED IS:

1. A pressure-sensitive adhesive comprising a
copolymer comprising on a copolymerized basis from about
5 55 to about 85% by weight of a monomer selected from the
group consisting of alkyl acrylate esters and alkyl
methacrylate esters containing from 4 to about 12 carbon
atoms in the alkyl group and mixtures thereof, from 0 to
about 35% by weight of an alkyl acrylate or methacrylate
10 ester containing less than 4 carbon atoms in the alkyl
group, from 0.01 to about 2% by weight of a glycidyl
monomer, from about 1 to about 10% by weight of an N-
vinyl lactam, and from 0 to about 15% by weight of an
unsaturated carboxylic acid, said copolymer having a
15 weight average molecular weight of at least about
200,000 and a glass transition temperature less than
about -15°C.

2. A pressure-sensitive adhesive as claimed in
20 claim 1 which contains, based on the total weight of
monomers, up to about 35% by weight, of a monomer
selected from the group consisting of polystyryl ethyl
methacrylate, aceto-acetoxy ethyl methacrylate, styrene,
alpha olefins, vinyl esters of alkanolic acids containing
25 more than about three carbon atoms and mixtures thereof.

3. A pressure-sensitive adhesive as claimed in
claim 1 in which the alkyl acrylate is 2-ethyl hexyl
acrylate or isooctyl acrylate.

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4. A pressure-sensitive adhesive as claimed in
claim 2 in which the alkyl acrylate is 2-ethyl hexyl
acrylate or isooctyl acrylate.

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1 5. A pressure-sensitive adhesive as claimed in
claim 1 in which the glycidyl monomer is selected from
the group consisting of glycidyl acrylate, glycidyl
methacrylate, allyl glycidyl ether and mixtures thereof.

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6. A pressure-sensitive adhesive as claimed in
claim 2 in which the glycidyl monomer is selected from
the group consisting of glycidyl acrylate, glycidyl
methacrylate, allyl glycidyl ether and mixtures thereof.

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7. A pressure-sensitive adhesive as claimed in
claim 3 in which the glycidyl monomer is selected from
the group consisting of glycidyl acrylate, glycidyl
methacrylate, allyl glycidyl ether and mixtures thereof.

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8. A pressure-sensitive adhesive as claimed in
claim 4 in which the glycidyl monomer is selected from
the group consisting of glycidyl acrylate, glycidyl
methacrylate, allyl glycidyl ether and mixtures thereof.

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9. A pressure-sensitive adhesive as claimed in
claim 1 in which the N-vinyl lactam is selected from the
group consisting of N-vinyl pyrrolidone, N-vinyl
caprolactam and mixtures thereof.

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10. A pressure-sensitive adhesive as claimed in
claim 2 in which the N-vinyl lactam is selected from the
group consisting of N-vinyl pyrrolidone, N-vinyl
caprolactam and mixtures thereof.

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11. A pressure-sensitive adhesive as claimed in
claim 3 in which the N-vinyl lactam is selected from the
group consisting of N-vinyl pyrrolidone, N-vinyl
caprolactam and mixtures thereof.

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1 12. A pressure-sensitive adhesive as claimed in
claim 4 in which the N-vinyl lactam is selected from the
group consisting of N-vinyl pyrrolidone, N-vinyl
caprolactam and mixtures thereof.

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13. A pressure-sensitive adhesive as claimed in
claim 5 in which the N-vinyl lactam is selected from the
group consisting of N-vinyl pyrrolidone, N-vinyl
caprolactam and mixtures thereof.

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14. A pressure-sensitive adhesive as claimed in
claim 6 in which the N-vinyl lactam is selected from the
group consisting of N-vinyl pyrrolidone, N-vinyl
caprolactam and mixtures thereof.

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15. A pressure-sensitive adhesive as claimed in
claim 7 in which the N-vinyl lactam is selected from the
group consisting of N-vinyl pyrrolidone, N-vinyl
caprolactam and mixtures thereof.

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16. A pressure-sensitive adhesive as claimed in
claim 8 in which the N-vinyl lactam is selected from the
group consisting of N-vinyl pyrrolidone, N-vinyl
caprolactam and mixtures thereof.

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17. A pressure-sensitive adhesive as claimed in
claim 1 in which the formed copolymer is cross-linked by
exposure to heat, ionic additive, actinic radiation or
electron beam radiation.

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1 18. A pressure-sensitive adhesive comprising a
copolymer comprising from about 55 to about 85% of an
alkyl acrylate ester selected from the group consisting
of isooctyl acrylate and 2-ethyl hexyl acrylate, from
5 about 0.01 to about 2% by weight glycidyl methacrylate,
from about 1 to about 10% of a N-vinyl lactam selected
from the group consisting of N-vinyl pyrrolidone and N-
vinyl caprolactam, from about 5 to 13% by weight
10 unsaturated carboxylic acid, and up to about 35% by
weight methyl acrylate, said polymer having weight
average molecular weight from about 200,000 to about
500,000 and a glass transition temperature less than
about -15°C.

15 19. A pressure-sensitive adhesive as claimed in
claim 18 in which the formed copolymer is cross-linked
by exposure to heat, ionic additive, actinic radiation
or electron beam radiation.

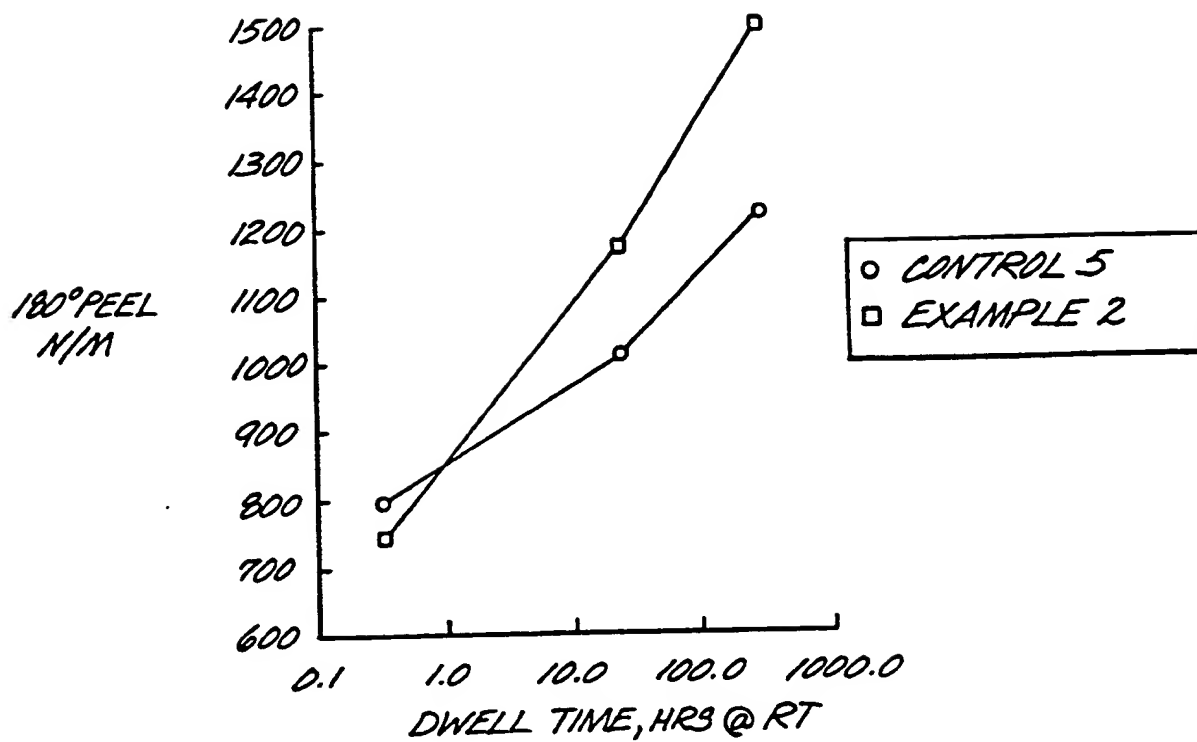
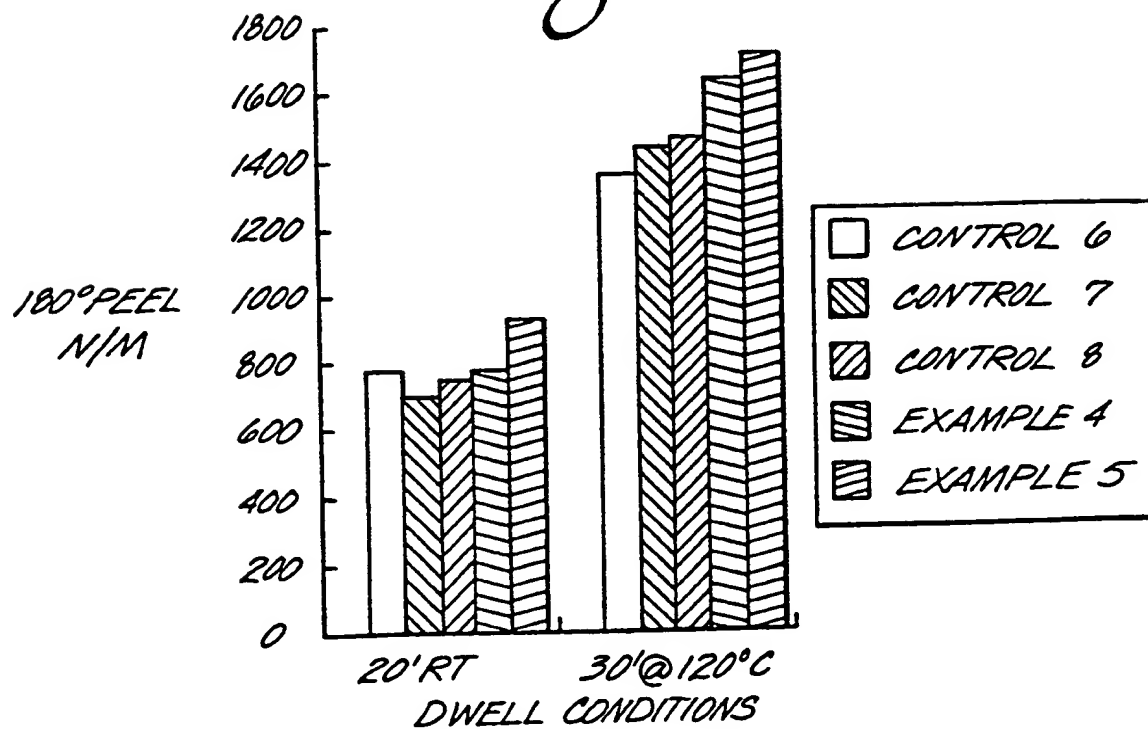
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Fig. 1*Fig. 2*

INTERNATIONAL SEARCH REPORT

International Application No. PCT/US88/04588

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) ⁶ According to International Patent Classification (IPC) or to both National Classification and IPC Int Cl ⁴ CO8F, 26/10; 24/00; USC1: 526/264,273											
II. FIELDS SEARCHED <div style="text-align: center; border-top: 1px solid black; border-bottom: 1px solid black; margin: 5px 0;">Minimum Documentation Searched ⁷</div> <table style="width: 100%; border-collapse: collapse;"> <tr> <th style="width: 25%; border: 1px solid black; padding: 5px;">Classification System</th> <th style="border: 1px solid black; padding: 5px;">Classification Symbols</th> </tr> <tr> <td style="border: 1px solid black; padding: 5px; text-align: center;">USC1</td> <td style="border: 1px solid black; padding: 5px; text-align: center;">526/264,273</td> </tr> </table> <div style="border: 1px solid black; padding: 5px; margin-top: 5px;"> Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched ⁸ </div>			Classification System	Classification Symbols	USC1	526/264,273					
Classification System	Classification Symbols										
USC1	526/264,273										
III. DOCUMENTS CONSIDERED TO BE RELEVANT ⁹ <table style="width: 100%; border-collapse: collapse;"> <tr> <th style="width: 10%; border: 1px solid black; padding: 5px;">Category [*]</th> <th style="border: 1px solid black; padding: 5px;">Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²</th> <th style="width: 15%; border: 1px solid black; padding: 5px;">Relevant to Claim No. ¹³</th> </tr> <tr> <td style="border: 1px solid black; padding: 5px; text-align: center;">A</td> <td style="border: 1px solid black; padding: 5px;">US, A 3,787,380, STAMBERGER 22 January 1974; See Entire Document</td> <td style="border: 1px solid black; padding: 5px; text-align: center;">1-19</td> </tr> <tr> <td style="border: 1px solid black; padding: 5px; text-align: center;">A</td> <td style="border: 1px solid black; padding: 5px;">US, A 4,510,197, SHAH 09 April 1985; See Entire Document</td> <td style="border: 1px solid black; padding: 5px; text-align: center;">1-19</td> </tr> </table>			Category [*]	Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³	A	US, A 3,787,380, STAMBERGER 22 January 1974; See Entire Document	1-19	A	US, A 4,510,197, SHAH 09 April 1985; See Entire Document	1-19
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<div style="display: flex; justify-content: space-between;"> <div style="width: 48%;"> <p>[*] Special categories of cited documents: ¹⁰</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 48%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"&" document member of the same patent family</p> </div> </div>											
IV. CERTIFICATION <table style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 50%; border: 1px solid black; padding: 5px;"> Date of the Actual Completion of the International Search 24 February 1989 </td> <td style="width: 50%; border: 1px solid black; padding: 5px;"> Date of Mailing of this International Search Report <div style="font-size: 1.2em; font-weight: bold;">18 APR 1989</div> </td> </tr> <tr> <td style="border: 1px solid black; padding: 5px;"> International Searching Authority ISA/US </td> <td style="border: 1px solid black; padding: 5px;"> Signature of Authorized Officer <div style="text-align: center; margin-top: 10px;"> ALEX. H. WALKER </div> </td> </tr> </table>			Date of the Actual Completion of the International Search 24 February 1989	Date of Mailing of this International Search Report <div style="font-size: 1.2em; font-weight: bold;">18 APR 1989</div>	International Searching Authority ISA/US	Signature of Authorized Officer <div style="text-align: center; margin-top: 10px;"> ALEX. H. WALKER </div>					
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